PERSPECTIVE

National Science Open
2: 20220042, 2023
https://doi.org/10.1360/nso/20220042

Chemistry

Harnessing electron spins with carbon materials for quantum information applications

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Received 23 July 2022; Revised 29 October 2022; Accepted 31 October 2022; Published online 11 January 2023

The implementation of information technology relies on functional chemicals and materials. Semiconductors have led to the rise of the electronic information era. As the next generation of information technology, how to implement quantum information processing has become an urgent task [1]. In order to carry superposition state and entanglement, systems, which are better isolated from environment, are designed to boost the quantum coherence time. For example, superconducting circuits, which are the leading candidate of quantum processor, uses the transmon design and the mK temperature condition to reduce the influence of environmental noise. Trapped ions are another well-developed system to encode quantum information, and the ultrahigh vacuum and laser-cooling enable ideal isolation from thermal perturbation.

In contrast to these relatively high-demanding setups, chemistry provides an affordable way of building the quantum system from the bottom up [2]. In particular, carbon-based spin materials, such as paramagnetic fullerene [3] NV^- centers in diamond [4] and bottom-up synthesized graphene [5], can satisfy the requirement of environmental noise isolation even at room temperature. The reasons are twofold. First, in carbon materials, the magnetic environment fluctuation is mitigated, because the predominantly abundant isotope of ^{12}C has no nuclear spin. Second, the electric decoherence is negligible, as the spin-electric coupling effect in the light elements of carbon is very small. Herein, we briefly envision the path of harnessing spins with carbon materials for quantum information applications.

Preparation of the carbon-based spin materials. Pristine carbon materials have closed-shell structures, so electron spin needs to be introduced intentionally (Figure 1A). The spin centers can be prepared via bottom-up synthetic approaches. For example, radicals linked by graphene nanoribbons [5] were synthesized. The benefit of the bottom-up synthesis is the high controllability of the spin centers, provided that the

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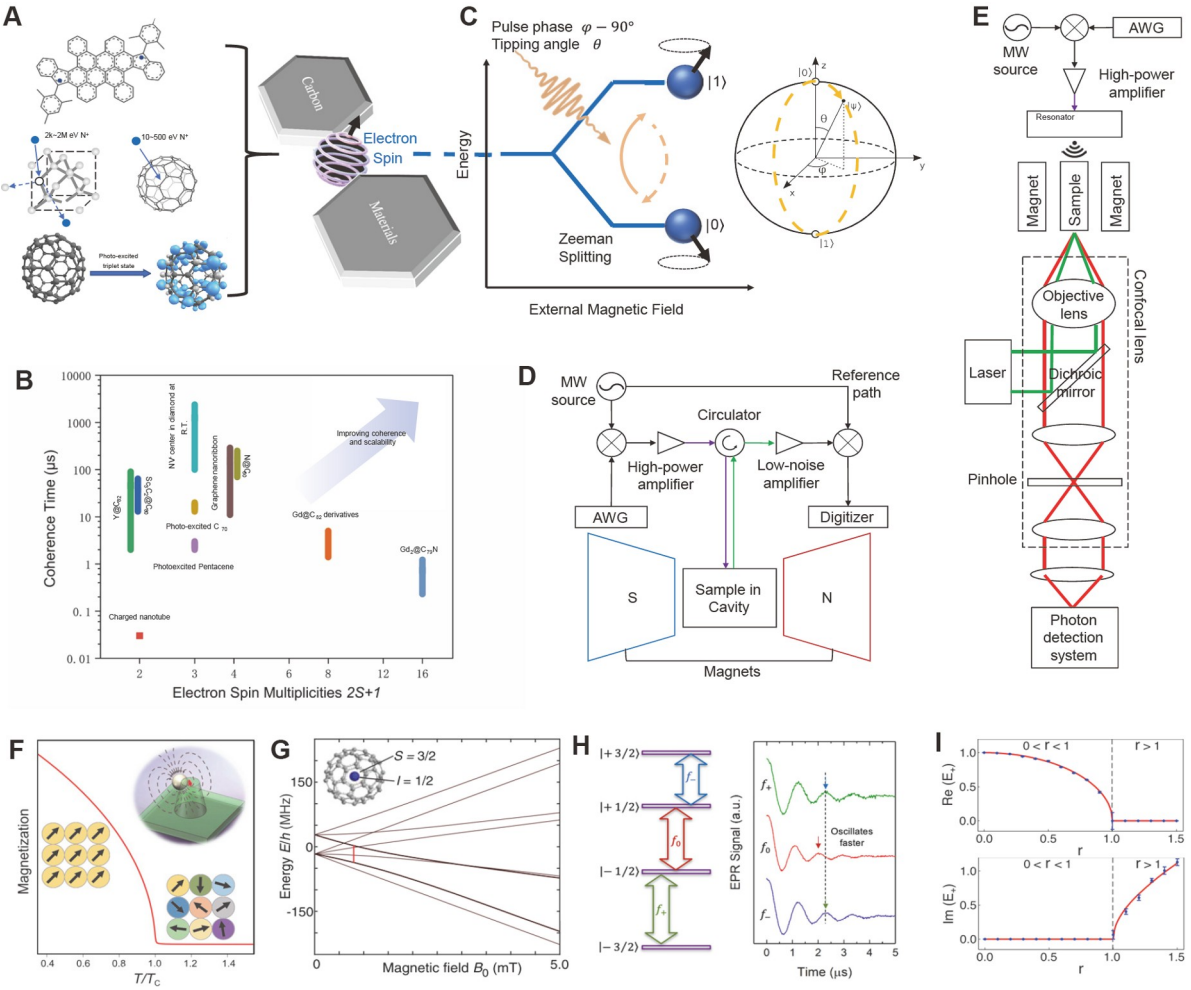

Figure 1 (A) Different ways to prepare an electron spin center in carbon materials. (B) Electron spin multiplicity and coherence time achieved with different types of carbon-based spin materials. (C) Schematic illustration of the quantum state of spin controlled by microwave pulses. (D) Instrument setups of EPR and (E) ODMR used to apply the spin manipulation and readout. (F) Ultra-sensitive quantum sensor for temperature detection. Reproduced with the permission of ref. [4]. (G) Solid-state atomic clock transitions for frequency standard. Reproduced with the permission of ref. [6]. (H) Addressable multi-level transitions to implement geomatic phase gate. Reproduced with the permission of ref. [3]. (I) Single quantum system experiment to manifest the break of parity-time symmetry with the r>1 region. Reproduced with the permission of ref. [7].

solubility challenges have been tackled. Yet, spin centers with the best quantum properties are obtained by ion implantation. For NV^- center in diamond, the ion beam is accelerated to relatively large energies ranging from keV to MeV [4], so that defects with certain penetration depth in the crystal would form. For endofullerenes, the ion beam energy is maintained very low, with a typical value of 40 eV, to prevent the delicate carbon cage structure from damaging [8]. The drawback of these protocols is the limited selectivity to yield the desired spin centers, therefore post-procedures of thermal treatment or chemical purification are needed, respectively. Photo radiation can be regarded as a special way of introducing a transient spin center. The photoexcited triplet states of pentacene and fullerene have been studied as qu*d*its [9]. In both systems, the triplet state is populated in a non-thermal equilibrated fashion, offering the spin polarization and strong spin signal that are hard to obtain otherwise. Nevertheless, due to limited intersystem crossing efficiency, the

scalability of transient spins is difficult to resolve.

Spin manipulation and readout. Depending on different structures of carbon-based spin materials, the electron spin multiplicity and coherence time vary (Figure 1B). Increased electron spin multiplicities can be used to encode higher-dimensional quantum information, and prolonged coherence times enable more quantum operations. Manipulation of the spin quantum state can be realized by magnetic resonance. Microwave pulse with the frequency that resonates with the energy gap triggers Rabi oscillation, so that arbitrary spin states on the Bloch sphere can be switched mutually on demand (Figure 1C). There are multiple ways to readout the spin state. Electron paramagnetic resonance (EPR) is designed to read the spin echo caused by the transverse magnetic momentum of the ensemble sample (Figure 1D). Optically detected magnetic resonance (ODMR) measures the intensity of the photoluminescence and resolve the population distribution of the spin state (Figure 1E). Benefiting from the high sensitivity of the photoluminescence signal, both ensemble spins and single spins can be measured using ODMR.

Quantum sensing applications. Due to the long coherence time of the spin center in carbon materials, the influence of surroundings can be sufficiently accumulated coherently, and then represented by the shifting or dispersion of the quantum phase of the spin. For example, by hybridizing NV^- center of diamond with a magnetic nanoparticle of copper-nickel alloy (Figure 1F), the resulting systems enable detection of 76 μK $Hz^{-1/2}$ around the Curie temperature of the alloy [4]. Because of the strong coupling and the high sensitivity, the temperature measurement is orders of magnitude improved than other techniques.

Solid-state molecular clocks. The spin system can also be tuned to an insusceptible condition, with which the idea of solid-state molecular clock has been proposed. Clock transition, a transition with a relatively stable frequency that is not affected by fluctuation of the external magnetic field, has been observed in endohedral fullerene (Figure 1G). Such property makes it a promising candidate toward solid-state molecular clock applications because all directions of magnetic field fluctuation can be effectively mitigated [6]. Compared with ammonia clock using gas cells, the solid-state design is advantageous for miniaturization.

Qubit and quantum computing applications. It has been studied by demonstrating different stages of functionality. Chemically functionalized graphene nanoribbon with radical centers have shown the effectiveness to encode qubit information with reliable Rabi oscillations [5]. Some fullerene species, such as photo-excited fullerene triplet state [9] and endohedral nitrogen fullerene derivatives [3], have been demonstrated with the ability of embedding qu*d*it (d=3 or 4), so that quantum phase interference and geomatic phase gates can be applied (Figure 1H). NV^- centers have achieved the furthest utility among all electron spin systems. While the fidelity of single quantum gate operations reaches >99.99%, two-qubit gates, multi electron spin entanglement, and Shor algorithm have also been demonstrated [10]. Moreover, quantum simulations were also enabled by the NV^- qubit utility such as demonstrating parity-time symmetry breaking [7] (Figure 1I).

From the synthesis of spin center to spin manipulation and applications, carbon materials have demonstrated advantages towards spin-based quantum information techniques. There is still a huge carbon material library waiting to be discovered with their spin bearing potential. Also, how to further scale-up the quantum dimension of the spin system, and how to efficiently polarize the spin system, are still open questions.

Funding

This work was supported by the National Natural Science Foundation of China (51802346 and 21822301), the National Key

R&D Program of China (2018YFA0306003 and 2021YFB3702304-4), and the National Science and Technology Major Project of China (J2019-VI-0017-0132).

Author contributions

Z.S., J.S.-D., and G.S. conceived the concept. C.H., W.Y.-X. and Z.S. wrote the manuscript. W.S. and M.W. contributed to the writing.

Conflict of interest

The authors declare that they have no conflict of interest.

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